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Acid Precipitation Patterns and Trends in Eastern North America, 1980-84

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STEVEN K. SEILKOP* AND PETER L. FINKELSTEIN**

U.S. Environmental Protection Agency, Research Triangle Park, NC 27711

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ABSTRACT

Using data compiled from seven nationwide precipitation chemistry networks in the United States and Canada, the spatial distribution of hydrogen, sulfate and nitrate ions in North America is discussed. Geographic patterns of concentration and deposition are characterized using isopleth maps which are presented and interpreted for the years 1980–84. The maps were developed using a geostatistical technique known as kriging. This method allows for the estimation of confidence limits on interpolated values. Using these limits, the significance of an observed decrease in the spatial patterns of deposition and concentration is assessed. Decreases of as large as 15–20% were observed in sulfate and nitrate concentrations between 1980 and 1983.

1. Introduction

Although there is widespread concern over the effects of acid precipitation, we have very little information about its past and present distribution over North America. For a few monitoring sites there is a long record of pH measurements, but in general we have only recently obtained enough data to characterize acid precipitation levels regionally. With the present monitoring network density in eastern North America, it is now possible to quantify regional patterns of concentration and deposition of sulfate, nitrate, and hydrogen ions in precipitation. In this paper, we investigate the spatial distribution of these constituents of acid precipitation for the 5 years 1980–84, the period in which chemical measurements of precipitation are available from a relatively large number of stations. Using a statistical technique known as kriging, our analysis provides an objective characterization of the geographic pattern of acid precipitation and utilizes the confidence limits on those patterns to assess whether there is evidence of significant changes in their amount and distribution over the 1980–84 time period.

2. Data

Data used in this study were gathered and summarized by the Acid Deposition System (ADS), an acid rain database operated by Pacific Northwest Laboratories for the U.S. Environmental Protection Agency (Olsen and Watson, 1984).

The database includes data from

- NADP (National Atmospheric Deposition Program)
- MAP3S/PCN (the MAP3S Precipitation Chemistry Network)
- CANSAP (Canadian Network for Sampling Precipitation)
- APN (The Canadian Air and Precipitation Monitoring Network)
- EPRI/SURE (EPRI Sulfate Regional Experiment Precipitation Chemistry Monitoring Program). Eight sites ceased operation in 1981; five sites became UAPSP sites.
- UAPSP (Utility Acid Precipitation Study Program)
- APIOS (Acid Precipitation in Ontario Study)

A brief summary of the deposition networks reported to the Acid Deposition System is given in Table 1. Each network has its own program for data quality assurance. In addition, ADS screens the data for reasonableness and completeness. We also checked the data a third time and removed a few outliers, as well as data from sites which the network operators felt were of lesser quality because of representativeness questions.

In this analysis only annual data are used. Concentrations are given as volume-weighted averages (milligrams per liter) while deposition represents a total over the year (grams per square meter). For each site, decisions had to be made as to whether or not there were enough individual samples to constitute a valid annual average. Four sampling representativeness criteria were used to screen stations for the analysis:

1) Percent Precipitation Coverage Length (PPCL): the percent of the days in the year for which valid precipitation measurements are available.

2) Percent Total Precipitation (PTP): the percent of

* Present affiliation: Analytical Sciences, Inc., Research Triangle Park, NC 27709.

** On assignment from NOAA, Dept. of Commerce.

TABLE 1. Description of monitoring networks.

Network	Geographical area	Number of sites as of 1982	Sampling period
NADP	National coverage of United States	117	Weekly
MAP3S/PCN	Eastern United States	9	Event
CANSAP	National coverage of Canada	62	Monthly
APN	Eastern Canada	9	Daily
APIOS C	Ontario province	36	Monthly
APIOS D	Ontario province	16	Daily
UAPSP	Eastern and central United States	19	Daily

the total precipitation that occurred during valid sampling (i.e., when good samples were taken) to the total measured precipitation. This figure would be low, for example, if there were significant rainfall while the samplers were inoperative.

3) Percent Valid Sampling Length (VSL): the percent of the year during which valid sampling could occur whether it rained or not. This is 100 minus the percent of the time the collector was known to be inoperative.

4) Station Representativeness: a subjective classification of stations made by the individual network operators, based upon exposure and distance to nearby sources of pollution.

In addition, a fifth criterion, Percent Collection Efficiency (PCE), was used to judge the quality of the sample. The PCE is the ratio expressed as a percent of the total sample volume for the year (expressed as a depth) to the total yearly depth measured in the recording raingage colocated with the collector. It is a measure of the quality of the site, the maintenance of the collector (and gage) and an indication of unusually high winds or snowfall which affect collector performance. For this study the criteria used to accept annual averages from a station are

PPCL > 90%
PTP > 75%
VSL > 75%
PCE 75% to 115%.

These criteria were selected in order to choose stations with enough data to characterize annual deposition or concentration adequately. The criteria were relatively stringent, resulting in the rejection of 40% of the stations in 1980, 25 to 35% in 1981–83, and 20% in 1984. Since we were most interested in accurately portraying annual spatial patterns, it was essential to eliminate values from sites with incomplete or misleading data.

These screening criteria were applied to annual data from eastern North America (east of 94 degrees west longitude), resulting in the following numbers of sites available for analysis:

Year	1980	1981	1982	1983	1984
Sites	43	70	96	101	62

The site locations are illustrated in Fig. 1. The relatively small number of monitoring stations depicted in 1984 is due to unavailability of data at this time (some cutoff date had to be made) rather than a reduction in monitoring activity. There are now more than 250 monitoring sites in North America. These maps clearly show the increases in monitoring activity since 1980, particularly that occurring in southern Ontario. They also demonstrate that the monitoring sites available for spatial analysis are quite irregularly spaced, with heavy sampling densities in some areas, e.g. eastern Ohio, and pockets of low density as seen in Maine and Quebec. The differential in sampling density has an impact on the geographic characterization and interpretation of the data from these sites. Clearly, we should have greater confidence in summary measures for areas represented by large numbers of sites than for those with sparse monitoring site coverage.

3. Analysis methodology

In investigating spatial and temporal trends in acid precipitation, it is desirable to have a visual means to represent geographic patterns. Isopleth maps were used to provide characterizations of concentration and wet deposition of sulfate, nitrate, and hydrogen ion over eastern North America for each of the years 1980–84. These maps are a powerful way to portray the general spatial features of the data but should be used judiciously. It should be understood that there is uncertainty (error) in the location of an isopleth due to random variations in the data on which it is based. Just as a sample mean imperfectly estimates the population mean, a group of isopleths imperfectly represents the true response surface associated with a spatial variable. Unfortunately, most mapping techniques for producing isopleths are incapable of reflecting the imprecision in the location of the isopleths due to their reliance on interpolation and smoothing algorithms which fail to quantify and utilize measures of data variability.

In order to redress these problems, an interpolation method known as kriging was used to provide a representation of the geographic pattern of acid precipitation. Unlike most other interpolation techniques, kriging uses the intersite correlation in the data to determine the weights that are applied to measured values in estimating unknown values. It also quantifies interpolation error, thereby providing a measure of the uncertainty in isopleth maps arising from kriging. Kriging is actually a family of "geostatistical" estimation procedures. It was named for a South African mining engineer, D. H. Krige, and has been used primarily in the mining industry. However, this methodology is be-

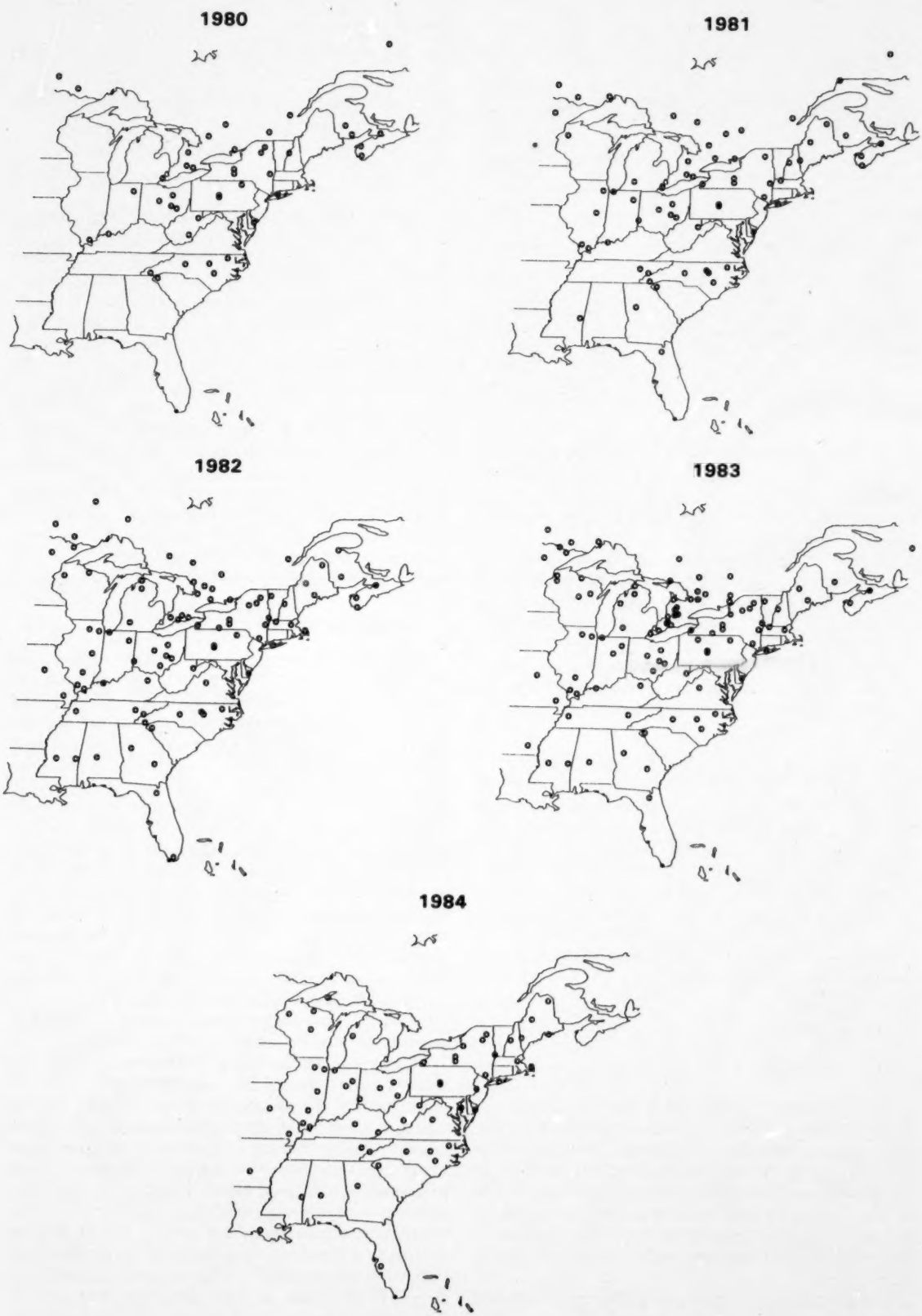

FIG. 1. Monitoring site locations, 1980–84.

coming increasingly popular in other fields (see Barnes, 1980, for descriptions). It has been found to be a particularly useful tool for analyzing spatial trends in acid precipitation data (Finkelstein and Seilkop, 1981; Eynon and Switzer, 1982; Finkelstein, 1984; Bilonick, 1983; and Seilkop, 1986).

All kriging techniques rely on a model of the spatially—and possibly temporally—related covariance structure of the data being analyzed. In simple kriging, which will be used in this paper, spatial variability is is modeled by the semivariogram, a function which relates the covariance of station-to-station differences to the distance between stations. A typical variogram is displayed in Fig. 2. Its positive slope reflects decreasing correlation between data as the distance between monitoring sites increases. The value of gamma at zero distance can be interpreted as the underlying error in the measurement process (Finkelstein, 1984). Several general models for variograms have been proposed (see Delhomme, 1979, for details). In this example, a curved line would be used.

Several forms of kriging (e.g., universal, generalized covariance) have been developed to remove the effect of data trends on the estimation of the spatial covariance structure. Simple kriging, however, is based on the assumption of a locally stable mean. Consequently, a variogram which contains some systematic (trend) components can lead to inflated estimates of interpolation error. However, the estimation bias is not serious under moderate spatial trends (Journel and Huijbregts, 1978). In an effort to understand the extent and effect of trends on variogram estimation, overall and directional (N–S, E–W, NE–SW, NW–SE) variograms were estimated for deposition and concentration of sulfate, nitrate, and hydrogen ion. In this analysis, the overall nondirectional variograms were employed in the calculation of all kriging interpolation weights and variances as the directional changes were not large enough to alter either pattern or variance estimates substantially. In all cases, either straight or curved variogram models were utilized.

In order to characterize acid precipitation, the available monitoring site data were used to obtain kriging estimates of values at 500 regularly spaced grid points across eastern North America. These estimated values define a relatively smooth surface which retains the primary features of the data. The variability in the location of isopleths was depicted with one standard deviation "confidence isopleths" which are derived using the kriging interpolation error variance estimates for each of the estimated grid values (see Finkelstein, 1984; Seilkop, 1986). Upper and lower confidence isopleths provide an envelope of uncertainty for a given isopleth. If areas bounded by these confidence isopleths fail to overlap across years, there is good evidence for a change in the spatial pattern of the variable being analyzed. A more appropriate assessment of the statistical significance of changes over time can be made by calculating the following ratio:

$$Z = \frac{Y_1 - Y_2}{[\mathrm{var}(Y_1) + \mathrm{var}(Y_2)]^{.5}} \qquad (1)$$

where Y_1 and Y_2 are estimated grid values for 2 yr, and $\mathrm{var}(Y_i)$, $i = 1, 2$ are the interpolation variances associated with these estimated values. Assuming that the difference is normally distributed, $|Z| > 1.96$ implies a significant difference at the 95% confidence level between the two years.

This is an extremely stringent test for changes in pattern because the variance of individual point esti-

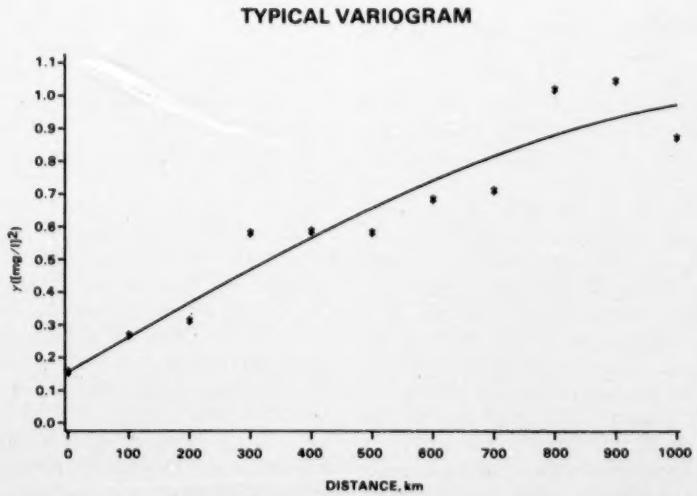

FIG. 2. Typical variogram for sulfate concentration, 1982. Asterisks are values estimated from data. The solid line represents the fitted spherical model.

mates will be large relative to the variability of the entire estimated surface. For a less severe test that provides better insight into temporal changes in spatial patterns, mean values in given areas were compared across years. Applying this methodology, we assess the significance of the change in level within that area over time. Again, kriging was used to estimate the areawide mean and its variance (Delhomme, 1978). Both the mean value and the variance were computed for areas 4° on a side throughout the study region. Because an areawide mean is based on several measurements, its variance is smaller than that of individual interpolated point value. Thus, comparisons of areawide averages from year to year are more sensitive than those made at individual points.

The choice of size and placement of the areas used in this analysis were arbitrary. The areas were centered on even steps proceeding in 4° steps from 30°N, 58°W. We found that these boundaries were a good compromise between variance reduction and spatial resolution. The results of this comparison are discussed in section 4.

4. Results

a. Isopleth maps

Isopleth maps developed using kriging are presented in Figs. 3–5 for sulfate deposition and concentration, nitrate deposition and concentration, and hydrogen ion deposition and concentration, respectively. Each figure contains maps for the years 1980–84. A few observations are obvious from the data. The response surfaces for all variables are reasonably well defined and are characterized by one major, fairly smooth pattern. In each map a clear central maximum can be seen, with decreasing values around it. Thus the existing sources and weather patterns do produce a definable area of maximum wet deposition.

The inconsistent sampling density across years, as well as the inherent variability in the estimated isopleths, diminishes our ability to make definitive statements regarding temporal trends in these data when comparing isopleth position alone. When evaluating changes in spatial patterns, it is important to consider the uncertainty in the estimation of the location of these isopleths. An example of the effect of increased sampling density on our confidence in individual estimates of constituents of acid precipitation is shown in Fig. 6. These maps depict the variances associated with estimated sulfate concentrations in 1980 and 1982. They reflect the uncertainty in the interpolation point estimates that were used to derive the maps shown in Fig. 3. The effect of the doubled sampling density for this period is clearly evident. The large increase in sampling activity in southern Ontario and the addition of sites in the United States reduce the point estimation variance in the area of maximum sulfate deposition by approximately 25%, considerably more than that in the South.

The size of the estimation variance for individual values is also quite striking. In a large part of the area of maximum deposition, the coefficient of variation is approximately 15% in 1982 and 20% in 1980. Clearly, isopleths derived from such variable estimates should be viewed with caution. As an example, consider the 3.5 mg/l sulfate concentration isopleths shown in Fig. 7. The solid lines are the estimated isopleths and the dashed lines represent the one standard deviation upper and lower confidence isopleths. An interpretation of the area bounded by the confidence isopleths is that approximately 70% of the isopleths resulting from a similar configuration of sampling sites would result in a 3.5 isopleth within this region. The shrinkage in size of this area between 1980 and 1982 accentuates the importance of using as much data as possible in defining spatial patterns. Given the uncertainty in the location of these isopleths, it is clear that assessments of temporal changes in isopleth patterns should be undertaken carefully. The uncertainty in the estimated isopleths should be taken into account and comparisons should be based on patterns with the least uncertainty, and consequently the maximum number of possible stations. Thus, in comparing annual spatial patterns, all data which passed the screening criteria were used. We found that application of these criteria resulted in substantial reduction in the uncertainty associated with estimated isopleths even though the number of sites used in the analysis was decreased. To further enhance our ability to discern temporal changes in patterns, we applied the areawide statistical analysis described above.

Considering the sulfate deposition maps first (Fig. 3), we see that isopleths generally have a SW to NE major axis, with a maximum centered between northern Ohio and southern Ontario in the vicinity of Lake Erie. It can be seen from Fig. 3 that there seems to be a general decrease in sulfate deposition between 1980 and 1983, with perhaps a slight increase in 1984. Both the peak level and the area of generally higher levels around it decreased about 15% in the 1980–83 period. Another way to consider this is to note that the area enclosed by an isopleth—for example, the 3.0 g m^{-2} isopleth—decreased substantially over this period. We will discuss the statistical significance of the changes later. Interestingly, the levels where there are measurements far away from the peak (which may arguably be considered quasi-background) do not seem to have changed much from year to year; this seems to be the case for all the other sets of maps, as well. With the exception of 1984, the sulfate concentration patterns are remarkably similar to the sulfate deposition patterns for all years. As with deposition, the decrease in concentration from 1980 to 1983 is quite evident. This consistency between concentration and deposition patterns is also evident for nitrogen and hydrogen and seems to indicate that variation in rainfall amounts over large areas may not have a significant effect on the spatial patterns.

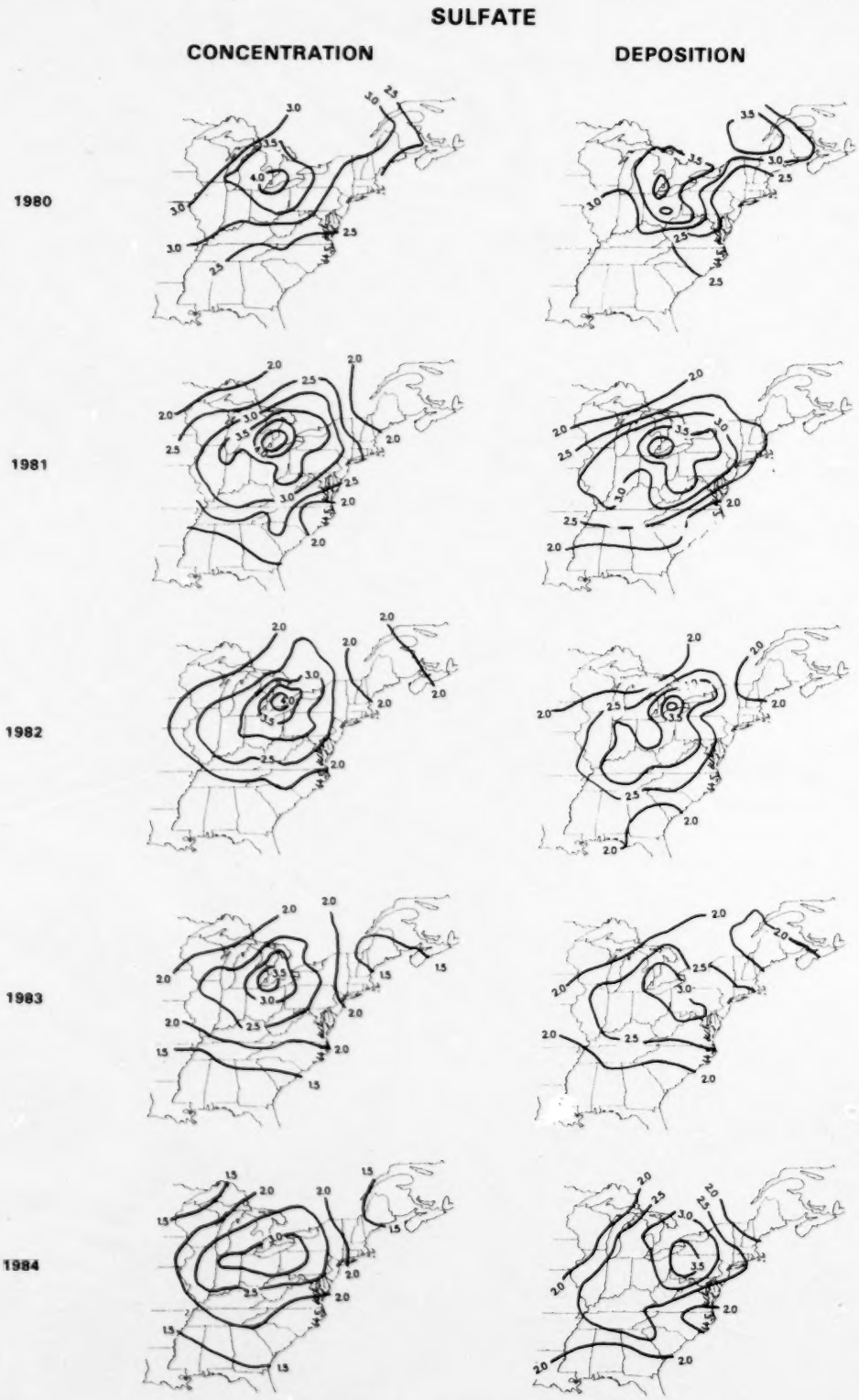

FIG. 3. Isopleth maps of average annual concentrations (mg l^{-1}) and total annual deposition (g m^{-2}) of sulfate in 1980–84.

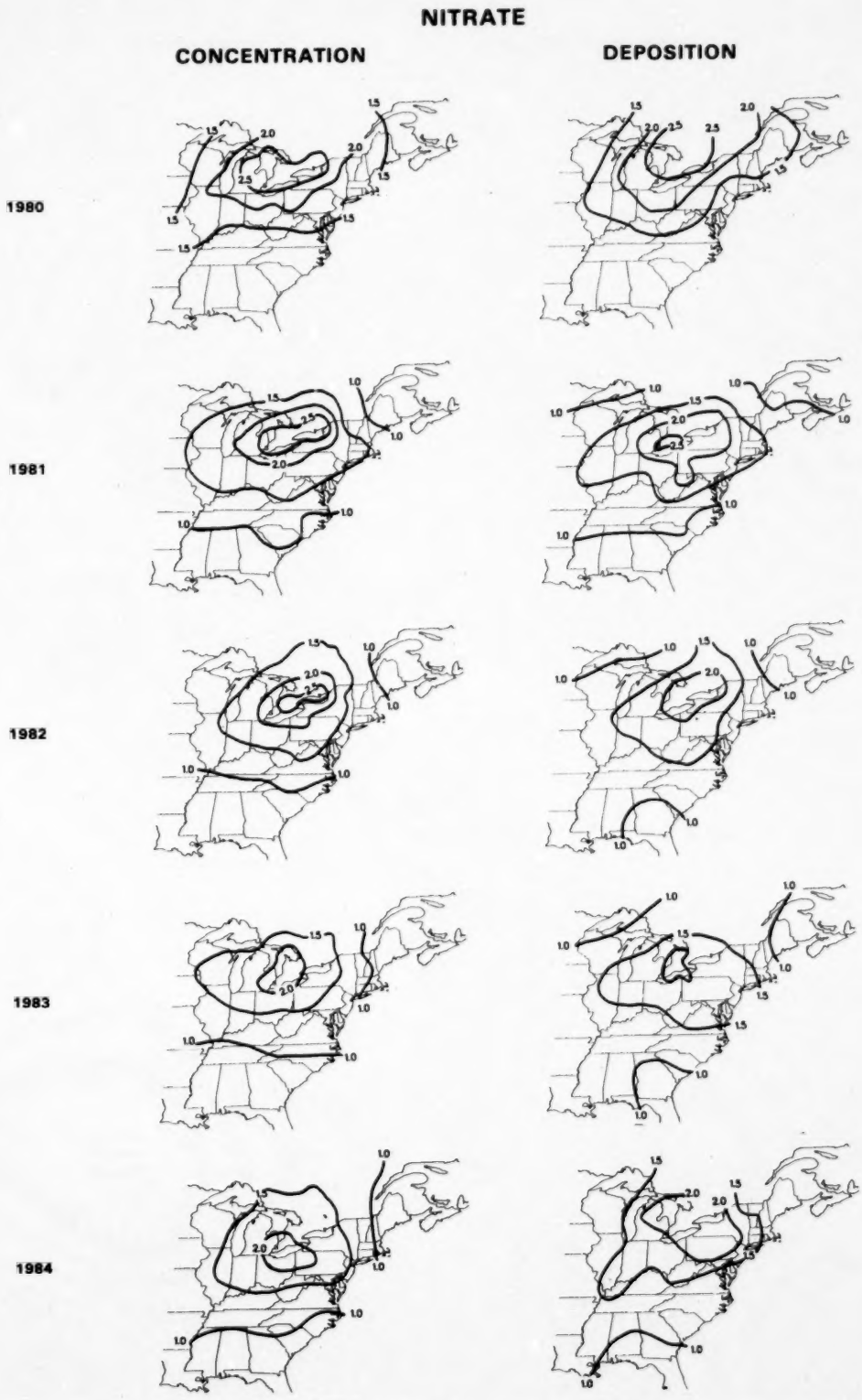

FIG. 4. Isopleth maps of average annual concentration (mg l^{-1}) and total annual deposition (g m^{-2}) of nitrate in 1980–84.

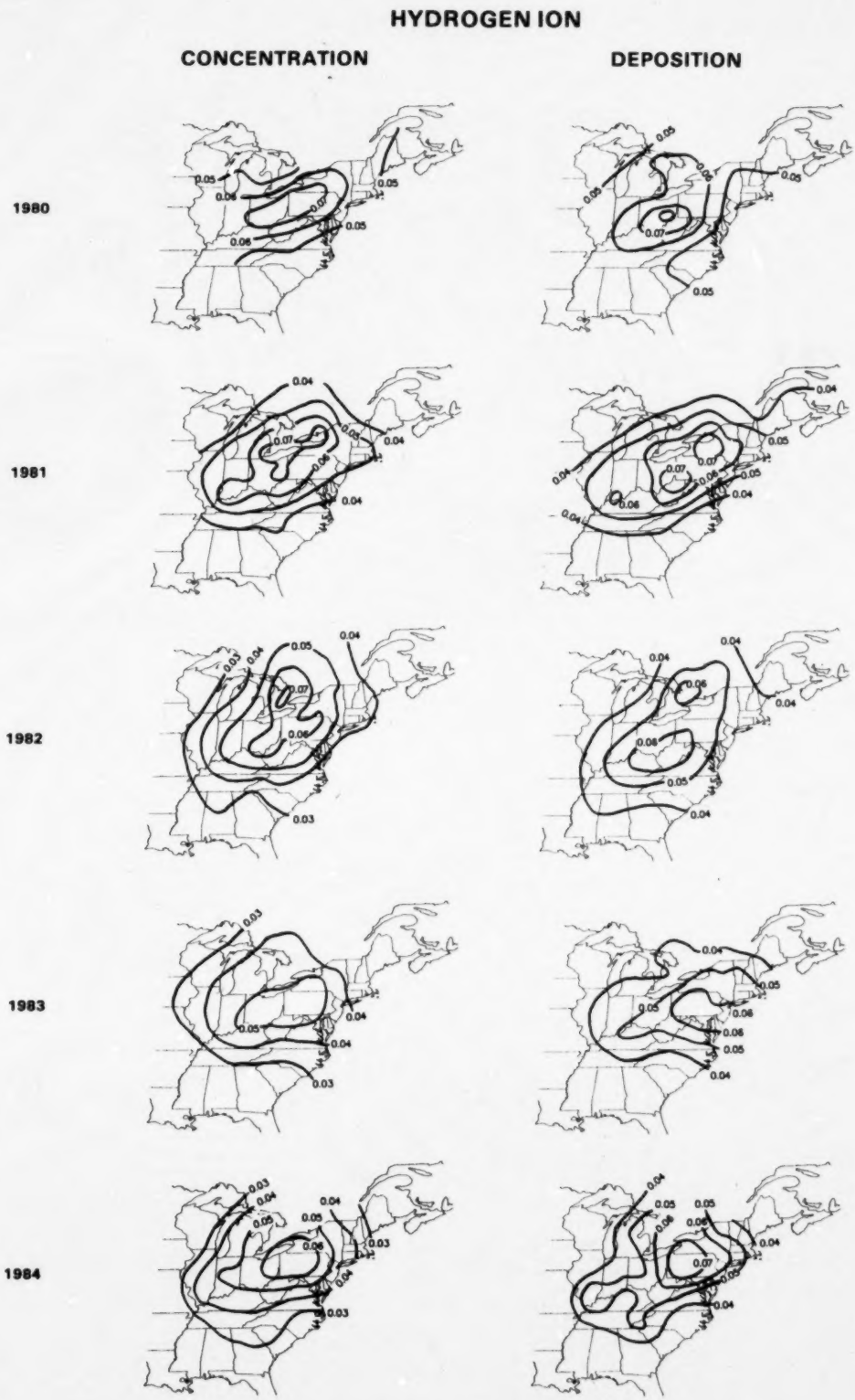

FIG. 5. Isopleth maps of average annual concentration (mg l^{-1}) and total annual deposition (g m^{-2}) of hydrogen ion in 1980–84.

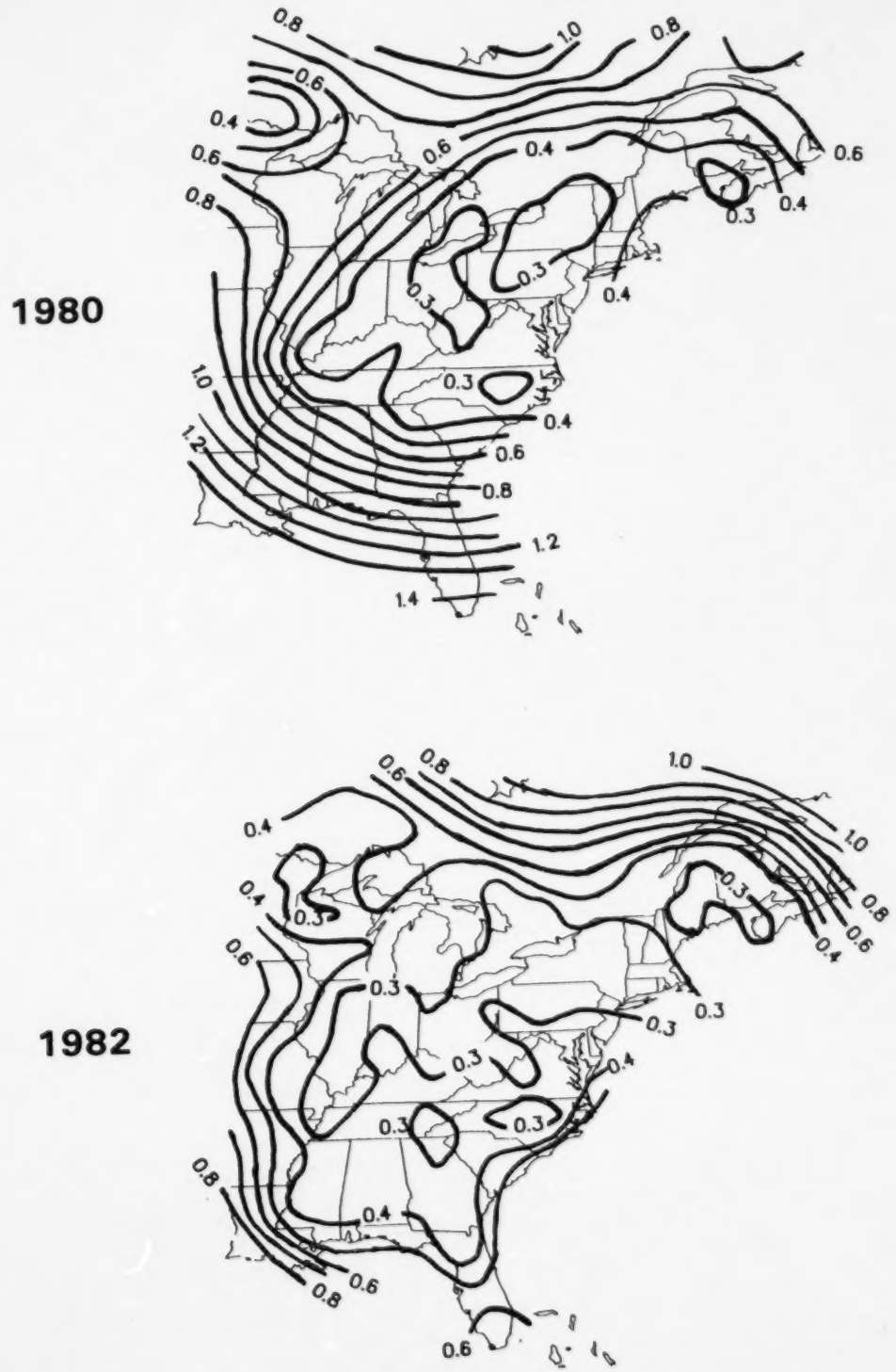

FIG. 6. Interpolation variances (mg l^{-1})^2 for kriging estimates of sulfate concentration in 1980 and 1982.

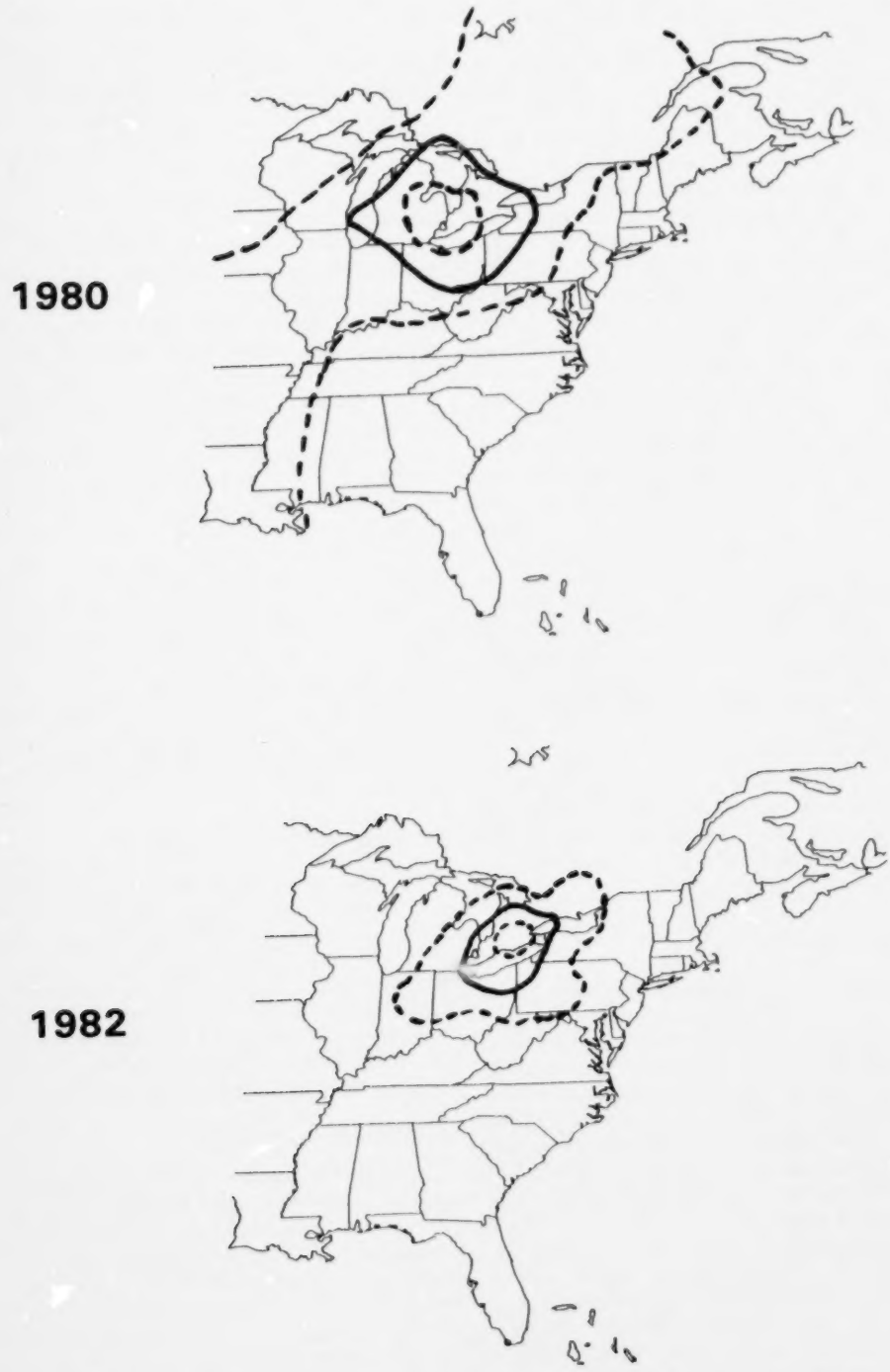

FIG. 7. Average annual 3.5 mg l^{-1} sulfate concentration isopleths for 1980 and 1982 (heavy lines) and their one standard deviation confidence limits (dashed lines).

The nitrate deposition patterns are, in general, quite similar to those of sulfate deposition, if perhaps a bit smoother. Over the 1980 to 1983 period, nitrate also shows a decrease of 15 to 20% in areas of high deposition and concentration. One noteworthy difference between the sulfate and nitrate patterns is that the nitrate maximum seems to be displaced slightly to the north and east. It is not at all clear that this is real, but it is somewhat consistent with the distribution of precursor emissions which also have a maximum farther to the east (Engineering Science, 1980).

The nitrate concentration maps again show similar patterns to the deposition maps, and a similar trend between 1980 and 1983. The isopleths are quite a bit smoother than those for sulfate concentration, perhaps reflecting the wide distribution of sources such as automobiles.

The hydrogen ion deposition maps would be expected to be very similar to those for sulfate and nitrate, and indeed they are. Perhaps it is a bit surprising that while their general shape and temporal trend is similar, the maxima appear to be shifted south and east of both the nitrate and sulfate maxima. The year 1983 especially seems to show a strong shift to the east which is not seen in either of the other maps. No explanation for this change has proven satisfactory at this time. We also note that the 1983 pattern has its major axis shifted more to an east–west orientation, similar to the sulfate pattern in this year.

Again, the hydrogen ion concentration maps are remarkably similar to those of hydrogen deposition, although the maximum is somewhat further north, and also looks quite similar to the sulfate concentration maps. The 1983 maximum does not show the shift to the east seen in deposition.

b. Comparison of area averages

As noted in section 3, estimates of the mean value of deposition and concentration over areas 4° of latitude and longitude on a side were compared for each year, along with the kriging estimate of the variance on the mean. The area averages for each year were compared to those in the chosen base year of 1981 to assess the statistical significance of the differences. Results of the comparison for sulfate for all years, and nitrate and hydrogen ion differences between 1981 and 1983 are shown in Figs. 8 and 9. In these figures, the shading depicts regions in which the area mean decreased significantly over the specified period.

The figures indicate that sulfate deposition decreased from 1980 to 1983 (i.e., 1980 to 1981, 1981 to 1982, and 1981 to 1983) in an area from southern Illinois through Ohio and western Pennsylvania to southern Ontario. This area corresponds rather closely to the area enclosed by the 3.0 g m^{-2} isopleth in 1981. There is evidence of decreases in sulfate concentration over most of the study area except the east and south coasts. Nitrate deposition decreased over a small area near the maximum of its pattern, which included northern Ohio, western Pennsylvania and New York, and south central Ontario. For nitrate concentration, the area of change was shifted to the east. Hydrogen ion deposition decreased only over a very small area near the center of its 1981 maximum, while the area of decrease for hydrogen ion concentration was quite wide, covering the Ohio River Valley from Illinois through western Pennsylvania and into western New England, Ontario and south through West Virginia and beyond. Changes for the other years (1980–81, 1981–82, 1981–84) were similar but smaller. The exact boundaries of significant change shown on these figures should not be interpreted too literally. A different choice of grid size or shape would have given slightly different results. Nonetheless, it is clear that during the period 1980–84 there were decreases in the deposition and concentration of species. One may deduce the location as well as magnitude of significant change in amount of wet deposition by considering these figures along with the isopleth maps.

An alternative strategy for identifying temporal trends is to evaluate changes at one or more individual stations over time rather than over a large area. This was successfully done by Schertz and Hirsch (1985). Their results also showed that when seasonal effects were removed, there was a decrease in deposition at several monitoring stations in the United States. Thus, our results are in qualitative agreement, and it would seem that independent approaches suggest this trend is real.

5. Discussion

The amount of wet deposition falling over an area is the result of a complex interaction of emission amount and location, transport, diffusion, atmospheric chemistry and precipitation. It is not the intent of this study to build a model which would account properly for all these variables; that is being done competently by others. We will, however, discuss two important variables which may help to explain the direction of the observed trends in acid precipitation.

a. Precipitation trends

One possible explanation for the changes in acid deposition levels from year to year is that significant changes may have taken place in the total amounts of precipitation. It is assumed that an increase in precipitation would cause an increase in deposition. To check this hypothesis, the total precipitation at each deposition monitoring site for each year was compared to the site's 5-yr mean total and to the long-term climatic mean at each site. To see if there were a trend in precipitation, the 5-yr mean at each site was subtracted from each year's total at the site, and the mean departure and standard error computed over all sites for each year. Only those sites which had at least 3-yr valid rain-

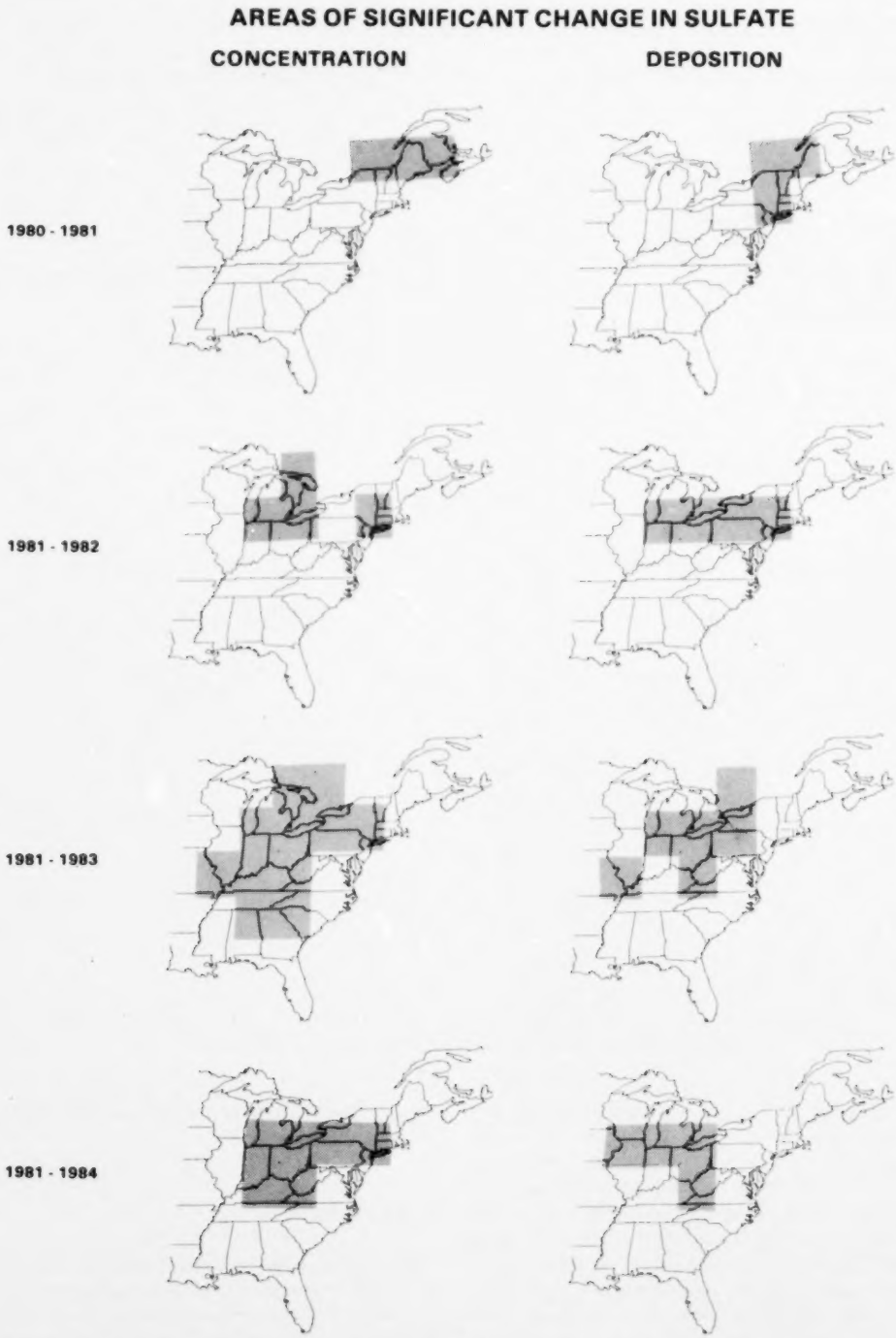

FIG. 8. Geographic areas of statistically significant ($p = 0.05$) decreases in sulfate concentration and deposition. Each map represents the decrease in the period indicated, e.g., between 1980 and 1981, 1981 and 1982, etc.

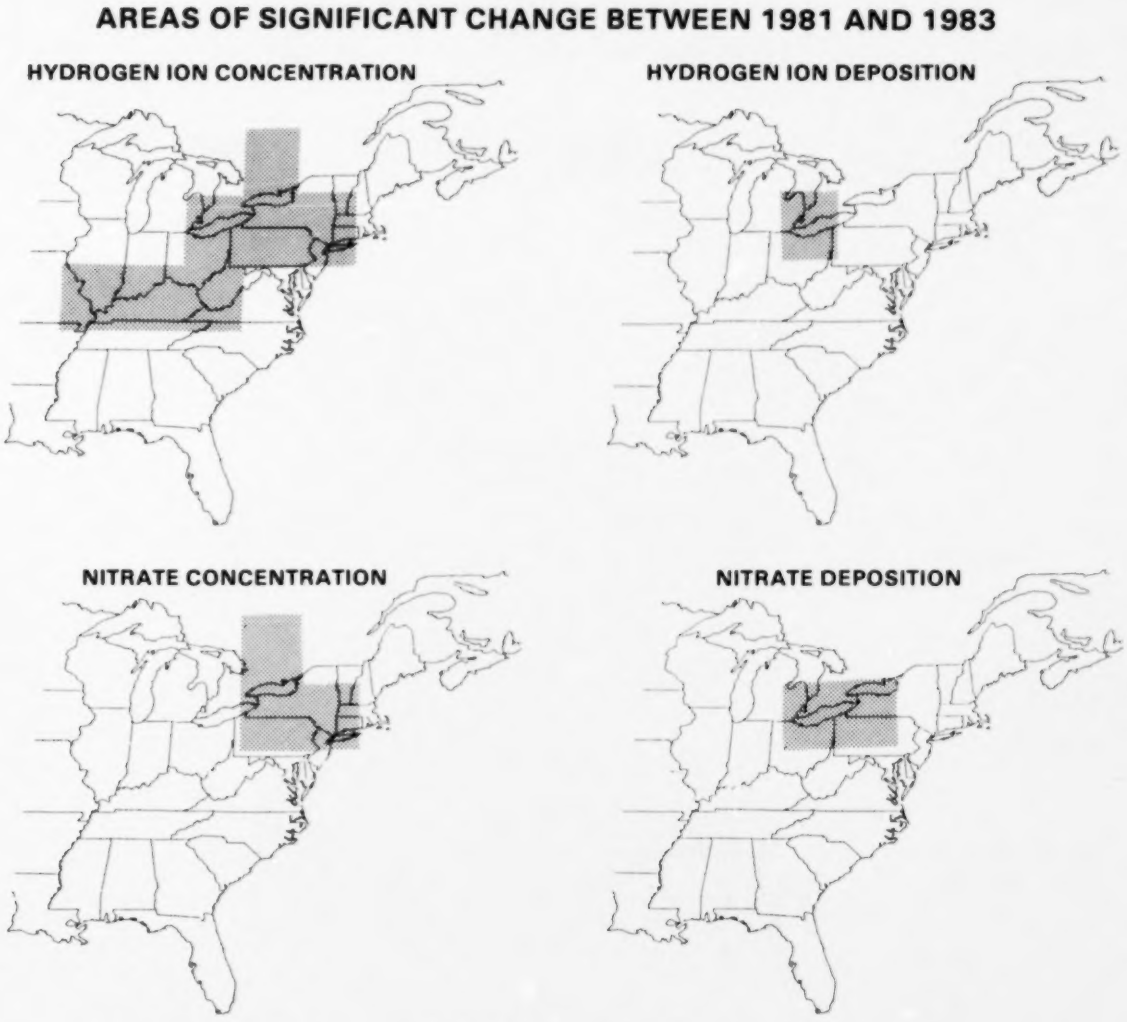

FIG. 9. Geographic areas of statistically significant ($p = 0.05$) decreases in annual average concentration and total deposition of nitrate and hydrogen ion between 1981 and 1983.

fall data were used. The results are shown in Fig. 10. The error bars are one standard error ($\sigma n^{-1/2}$). A significant trend from drier years in 1980 and 1981 to a wetter year in 1983 is clearly evident. The same analysis was performed for the area of maximum deposition (defined arbitrarily as 38° to 46°N, 74° to 86°W) and the surrounding area of low deposition. The results were similar, indicating that the amount of precipitation in the high-deposition areas also increased. This increase in precipitation may have had an impact on concentration levels by dilution, but since both concentration and deposition decreased, and by roughly the same amount, it is not evident why or how an increase in rainfall might cause such a decrease. It would seem that the changes in deposition occurred in spite of the changes in precipitation and not because of them.

b. Emissions

Another possible reason for trends in deposition may be trends in emissions. Annual emissions of SO_x and NO_x were examined to see if they could help explain the changes observed in acid rain levels. A report issued by EPA's Monitoring and Data Analysis Division (1986) estimates annual emissions from all sources in the United States for the years 1940–84. Their figures for the years 1979–84 are given in Table 2. These data suggest a decrease of about 10% for SO_x and 5% for

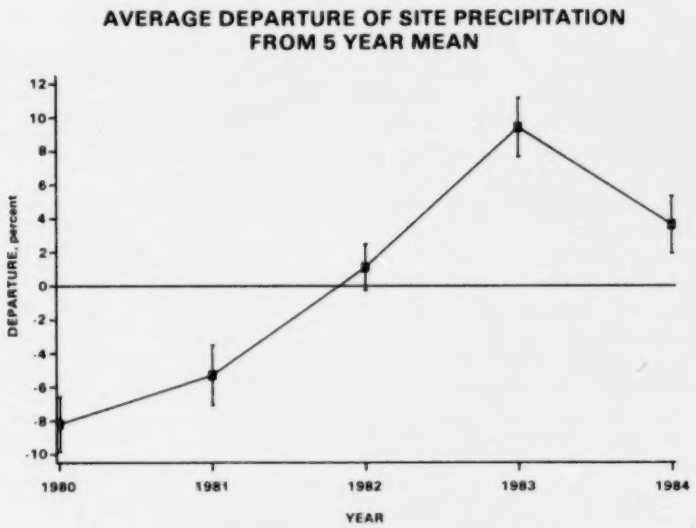

FIG. 10. Average percentage departure of precipitation at all sites from 5-yr average (1980–84).

NO_x up to 1983, with a slight increase in 1984. The relative 95% confidence limits on the emissions figures are thought to be at approximately ±5% (Mann, personal communication, 1987). After accounting for this level of measurement uncertainty, regression analysis results indicate that the observed trend is not likely to be attributable to chance alone ($p < 0.05$). The EPA report indicates that most of the SO_x emissions (~80%) come from stationary source fuel combustion, with a large part of the remainder coming from industrial processes. The NO_x emissions are approximately equally divided between stationary source fuel combustion and transportation related sources. Interestingly, the decrease for each category of NO_x emissions was about the same.

Knudson (1985) gives SO_x emissions by state for 1975–83. Emission figures from four states in the high-deposition area, which together account for approximately one-fourth of the total United States emissions, are given in Table 3. Note that while the trend of the four states together is similar to the national trend, there is quite a bit of variability from state to state.

These changes in emissions seem to be reflected in, and help to understand, the changes in wet deposition and concentration fields. Certainly the trends are in a similar direction and the results are not counterintuitive. It is also clear that no simple quantitative relationship between national emissions and large-scale changes in wet deposition is suggested or implied by this. The ratio of changes in emissions to deposition or concentration trends are different for different areas, and certainly different for each species. Further understanding of the complex source–receptor relationships requires detailed chemical, transport, and diffusion models.

6. Summary and conclusions

Over the years 1980–84, there is evidence of a 15–20% decrease in the total deposition and average concentration levels of sulfate, nitrate, and hydrogen ions in precipitation falling over eastern North America. These decreases have occurred in the geographic regions of maximum deposition and were most dramatic between 1981 and 1983. In 1984, there is some evi-

TABLE 2. Annual U.S. emissions in teragrams (10^{12} grams) per year.

Year	SO_x	NO_x
1979	24.5	21.1
1980	23.2	20.4
1981	22.3	20.5
1982	21.3	19.7
1983	20.6	19.1
1984	21.4	19.7

TABLE 3. Annual emissions of SO_x from four states in 1980–83 (measured in teragrams per year).

Year	Ill.	N.Y.	Ohio	Pa.	Total	% change from 1980
1980	1.33	0.78	2.45	1.58	6.14	0.00
1981	1.21	0.79	2.49	1.42	5.91	3.7
1982	1.23	0.74	2.42	1.37	5.76	6.2
1983	1.26	0.70	2.35	1.46	5.77	6.0

dence of increases in the levels of the primary constituents of acid precipitation, although they were still decreased relative to the levels observed in 1980 and 1981. Although the observed decreases correspond in direction to reported reductions in United States emissions of SO_x and NO_x, the emission figures are subject to estimation error and should be used cautiously. Variation in the physical processes involved must also be considered as an explanatory factor in the observed trends in acid precipitation. During the 1980–84 period, there is evidence of an increase in precipitation over eastern North America. This might explain the observed decrease in concentration of sulfate, nitrate, and hydrogen ions, but is not consistent with the decreases in deposition of these acidic precipitation constituents. The absence of a precipitation-based explanation for deposition decreases lends some strength to the plausibility of a relationship between the observed decreases in acid precipitation parameters and the reported reductions in emissions. However, the evidence is at this stage circumstantial, insofar as there are many other meteorological sources of variation that could affect the annual average concentration and deposition of pollutants in precipitation.

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